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Polarization dependence of resonance enhanced two-photon dissociation of diatoms for linear and circular polarizations

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Abstract : Polarization dependence of resonance enhanced two-photon dissociation of diatoms has been studied for one linear and the other circular polarizations of the two photon fields. It is found that the results strongly depend on whether the linear polarization is parallel or perpendicular to the direction of propagation of the circularly polarized photon For both parallel and perpendicular polarizations of the linearly polarized photon, the results are the same for linear + circular and circular + linear polarizations.

Keywords : Multiphoton processes, two-photon dissociation, polarization effect

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Previously [1] we have studied weak field resonance enhanced two-photon dissociation (RETPD) of H_2 by X $\rightarrow$ B(C) $\rightarrow$ EF + GK + I transitions with the polarizations of both the fields linear (parallel or crossed) or circular (same- or opposite-sense) for different values of rotational quantum numbers of the states involved using the reduced density operator formalism [2]. The angular momentum and polarization dependent factors entering the cross sections have been tabulated in our earlier works [1]. For bound states, as discussed by Wang and Xia [3], a linearly or circularly polarized pump laser can be used with a linearly polarized probe laser to determine the angular momenta of the excited molecular states. This technique, termed two-step polarization labelling spectroscopy, requires the formulae for relative intensities of the transitions for different polarizations.

Here we have studied the relative RETPD cross sections when one of the photons is linearly and the other circularly polarized for two different relative orientations (parallel and perpendicular) of their directions of propagation. Four distinct cases can be identified depending on whether the resonant exiciting photon (the first photon) or the dissociating photon (the second photon) is circularly polarized, and whether the polarization direction of the linearly polarized photon is parallel or perpendicular to the direction of propagation of the circularly polarized photon which is taken as the space fixed Z-axis. This second distinction applies only to photon beams propagating at right angles to each other since for colinear propagation, the polarization direction of the linearly polarized photon is necessarily perpendicular to the space fixed Z-axis. This can be understood with reference to Figure 1 where the geometry of the experimental arrangement is shown for the two cases. Since the first photon creates an oriented/aligned population in the resonant state, the dissociation in different channels will depend on the polarization of the second photon which is perpendicular to the Z-axis for colinear propagation but may be parallel or perpendicular for crossed propagation of the two photon beams. Thus, the branching ratio between final electronic states with different values of Λ (the electronic angular momentum projection quantum number) and for different values of J in the same Λ-state may be different in colinear and crossed configurations for one linearly and one circularly polarized beam. We have investigated four special cases, *viz.*, (i) the first photon polarization linear parallel to Z-axis and the second photon polarization circular (Figure 1(a)), (ii) the first photon polarization

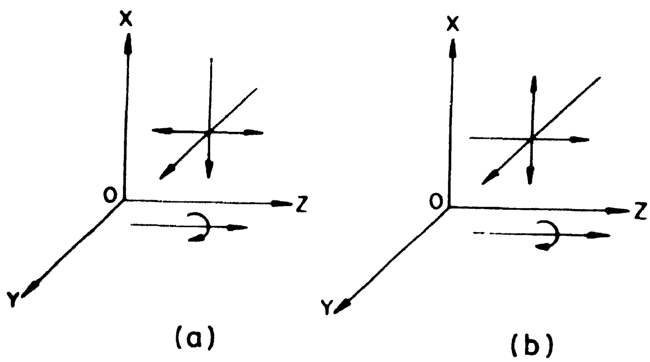

Figure 1. (a) One photon field of linear parallel and the other of circular polarizations, (b) one photon field of linear perpendicular and the other of circular polarizations.

linear perpendicular to Z-axis and the second photon polarization circular (Figure 1(b)), (iii) the first photon polarization circular and the second photon polarization linear parallel (Figure 1(a)), (iv) the first photon polarization circular and the second photon polarization linear perpendicular (Figure 1(b)). In cases (i) and (iii) the pump and probe beams are crossed, whereas in cases (ii) and (iv) the pump and probe beams may be colinear or crossed in respect of propagation. For each set of polarizations, we have calculated the geometry determining angular factors for initial Σ, intermediate Σ/Π and final Σ/Π states. It is found that the cases (i) and (iii) are equivalent and so are the cases (ii) and (iv).

For one photon linearly and the other circularly polarized the specific form of the angular factor of the RETPD cross section [1] is

$$A_{J_g J_i J_f}^{\Lambda_g \Lambda_i \Lambda_f} = \left(2J_i+1\right)\left(2J_f+1\right)\left\{S_1^{\Lambda_g \Lambda_i} S_2^{\Lambda_i \Lambda_f} \big/ R_1^{\Lambda_g \Lambda_i} R_2^{\Lambda_i \Lambda_f}\right\}$$

$$\times\left[1/9 + (1/6)(-2)^{\alpha}\,(-1)^{J_f - J_g}\,(2J_i+1)\left(\begin{Bmatrix} 1 & 1 & 2 \\ J_i & J_i & J_g \end{Bmatrix}\right.\right.$$

$$\left.\left.\times\begin{Bmatrix} 1 & 1 & 2 \\ J_i & J_i & J_f \end{Bmatrix}\right)\right] \tag{1}$$

where $\{\because\}$ are $6j$ coefficients [4]. $\alpha = 1$ or 0 for the linear polarization parallel or perpendicular to the Z-axis. For parallel polarization of the linearly polarized photon ($\alpha = 1$), eq. (1) is exactly the same as the case of two linear perpendicular polarizations [1]. The factor $\left\{S_1^{\Lambda_g \Lambda_i} S_2^{\Lambda_i \Lambda_f} \big/ R_1^{\Lambda_g \Lambda_i} R_2^{\Lambda_i \Lambda_f}\right\}$ defined elsewhere [1] depends on the electronic transitions involved. The calculated values of the angular factors for parallel ($\alpha = 1$) and perpendicular ($\alpha = 0$) polarizations of the linearly polarized photon are given in Tables 1(a) and 1(b), respectively. Tanaka and Kawasaki [5] studied the two-photon absorption for linearly and

Table 1(a). Angular factors $A_{J_g J_i J_f}^{\Lambda_g \Lambda_i \Lambda_f}$ for RETPD with linear parallel and circular polarizations of the two photon fields.

J_g	J_i	J_f	$A_{J_g J_i J_f}^{0,0,0}$	$A_{J_g J_i J_f}^{0,\pm1,0}$	$A_{J_g J_i J_f}^{0,0,\pm1}$	$A_{J_g J_i J_f}^{0,\pm1,\pm1}$
0	1	0	–	–	–	–
		1	–	–	1/6	1/6
		2	1/15	1/15	1/10	1/10
1	0	1	1/27	–	2/27	–
1	1	0	–	–	–	–
		1	–	1/12	–	1/24
		2	–	–	–	7/120
1	2	1	13/675	13/300	13/1350	13/600
		2	–	–	1/10	1/40
		3	1/25	1/25	4/75	4/75
2	1	0	1/75	1/75	–	–
		1	–	–	7/150	7/600
		2	11/375	11/1500	11/250	11/1000

Table 1(a). (*Cont'd*)

J_g	J_i	J_f	$A^{0,0,0}_{J_gJ_iJ_f}$	$A^{0,\pm1,0}_{J_gJ_iJ_f}$	$A^{0,0,\pm1}_{J_gJ_iJ_f}$	$A^{0,\pm1,\pm1}_{J_gJ_iJ_f}$
2	2	1	–	–	–	9/200
		2	–	13/180	–	13/1080
		3	–	–	–	44/675
2	3	2	19/875	304/7875	38/2625	608/23625
		3	–	–	13/150	13/1350
		4	6/175	6/175	3/70	3/70
3	2	1	3/175	3/175	3/350	3/350
		2	–	–	11/210	11/1890
		3	34/1225	136/11025	136/3675	544/33075
3	3	2	–	–	–	52/945
		3	–	5/72	–	5/864
		4	–	–	–	15/224
4	3	2	2/105	2/105	4/315	4/315
		3	–	–	1/18	1/288
		4	46/1701	23/1512	115/3402	115/6048

Table 1(b). Angular factors $A^{\Lambda_g\Lambda_i\Lambda_f}_{J_gJ_iJ_f}$ for RETPD with linear perpendicular and circula polarizations of the two photon fields.

J_g	J_i	J_f	$A^{0,0,0}_{J_gJ_iJ_f}$	$A^{0,\pm1,0}_{J_gJ_iJ_f}$	$A^{0,0,\pm1}_{J_gJ_iJ_f}$	$A^{0,\pm1,\pm1}_{J_gJ_iJ_f}$
0	1	0	1/18	2/9	–	–
		1	–	–	1/12	1/12
		2	7/90	7/90	7/60	7/60
1	0	1	1/27	–	2/27	–
1	1	0	–	–	–	–
		1	–	1/8	–	1/16
		2	–	–	–	13/240
1	2	1	47/1350	47/600	47/2700	47/1200
		2	–	–	11/180	11/720
		3	7/150	7/150	14/225	14/225
2	1	0	7/450	7/450	–	–
		1	–	–	13/300	13/1200
		2	67/2250	67/9000	67/1500	67/6000
2	2	1	–	–	–	11/400
		2	–	47/360	–	47/2160
		3	–	–	–	38/675
2	3	2	4/125	64/1125	8/375	128/3375
		3	–	–	17/300	17/2700
		4	1/25	1/25	1/20	1/20

Table 1(b). (*Cont'd*)

J_g	J_i	J_f	$A^{0,0,0}_{J_gJ_iJ_f}$	$A^{0,\pm1,0}_{J_gJ_iJ_f}$	$A^{0,0,\pm1}_{J_gJ_iJ_f}$	$A^{0,\pm1,\pm1}_{J_gJ_iJ_f}$
3	2	1	1/50	1/50	1/100	1/100
		2	–	–	19/420	19/3780
		3	71/2450	142/11025	142/3675	568/33075
3	3	2	–	–	–	34/945
		3	–	19/144	–	19/1728
		4	–	–	–	24/448
4	3	2	1/45	1/45	2/135	2/135
		3	–	–	5/108	5/1728
		4	7/243	7/432	35/972	35/1728

circularly polarized light propagating along the same direction. They have calculated the polarization ratios for $J_g = 0$, $J_i = 1$, and $J_f = 2$ with $\Lambda_g = \Lambda_i = \Lambda_f = 0$ only. Their results agree with the values calculated from our expressions of the angular factors. From the calculated values of the angular factors for one linear and the other circular, it is concluded that the RETPD cross sections [1] of diatoms will strongly depend on whether the linear polarization is parallel or perpendicular to the direction of propagation of the circularly polarized photon.

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